



IN THE UNITED STATES PATENT & TRADEMARK OFFICE

In re Application of

Bernd Zäschke et al.

Serial Number 10,507,315

Filed: 09/10/2004

For: GRAFT POLYOLS WITH A BIMODAL PARTICLE SIZE DISTRIBUTION AND
METHOD FOR PRODUCING GRAFT POLYOLS OF THIS TYPE, IN ADDITION TO
THE USE THEREOF FOR PRODUCING POLYURETHANES

D E C L A R A T I O N U N D E R 37 C F R 1.132

I, Daniel Freidank, a citizen of the Federal Republic of Germany and residing at
67063 Ludwigshafen, Federal Republic of Germany, declare as follows:

I am a fully trained chemist, having studied chemistry at the University of Bielefeld, 33501
Bielefeld, Federal Republic of Germany, from 1993 to 1999 and at the University of Freiburg,
79110 Freiburg i. Br., Federal Republic of Germany, from 2000 to 2003.

I received my Doctors degree at the University of Freiburg in 2005,

I joined BASF AG, 67056 Ludwigshafen, Federal Republic of Germany, in 2003, since when
I have been working in the field of polyurethane research and development,

I am well acquainted with technical English,

To demonstrate that the graft polyols prepared according to the above patent application have a bimodal particle size distributions with two separate peaks, that do not overlap, I made the following examples.

Comparative Experiment

Six samples of graft polyols had been prepared in accordance to Example 4 of the above patent application.

The particle size distribution of the graft polyols was measured by "laser diffraction analysis" in combination with "polarization intensity differential scattering (PIDS)". This method for the determination of particle size distribution is an important standard method in order to characterize dispersions. The companies Coulter and Malvern are the two major suppliers of instruments for particle size analysis based on laser diffraction.

The particle size distribution of a graft polyol sample according to the claims of WO 03/078496 was determined with a Laser Diffraction Particle Size Analyzer from Beckman Coulter GmbH Krefeld (www.beckmancoulter.com) under the following conditions:

Instrument:

Name:	Laser Diffraction Particle Size Analyzer LS 230
Dynamic Size Range:	0,04 μm up to 2000 μm
Measurement Principle:	Fraunhofer theory of light scattering in combination with Polarization Intensity Differential Scattering (PIDS)
Laser wavelength:	750 nm
PIDS light source:	Tungsten-Halogen-lamp 3 filters for polarization in the vertical and horizontal planes (450 nm; 600 nm; 900 nm) Measurement of light scattering at six scattering angles (60°; 75°; 90°; 105°; 120° and 146°)
Dispersion unit:	Small Volume Module (SVM-Module)
Detector:	132 detectors; 116 size channels
Software:	LS32 Version 3.01 (Microsoft Windows based)

Evaluation:

Optical model:	PSL (Polystyren Latex) with PIDS; Formfactor 1
Measurement liquid:	2-Propanol
Liquid refraction index:	1,374
Real part of refraction index of the sample:	1,6
Imaginary part of the refraction index of the sample:	0,0

Sample preparation:

0,5 ml Graftpolyol are added with a Pasteur pipette to 15 ml Isopropanol (2-Propanol) in a 30 ml-beaker. The components are well mixed. The dispersion is introduced drop wise into the dispersion unit of the measurement instrument. Sample introduction is finished when the instrument signalizes adequate density. Then the measurement can be started.

The results of the measurement are displayed graphically and as table

Picture 1

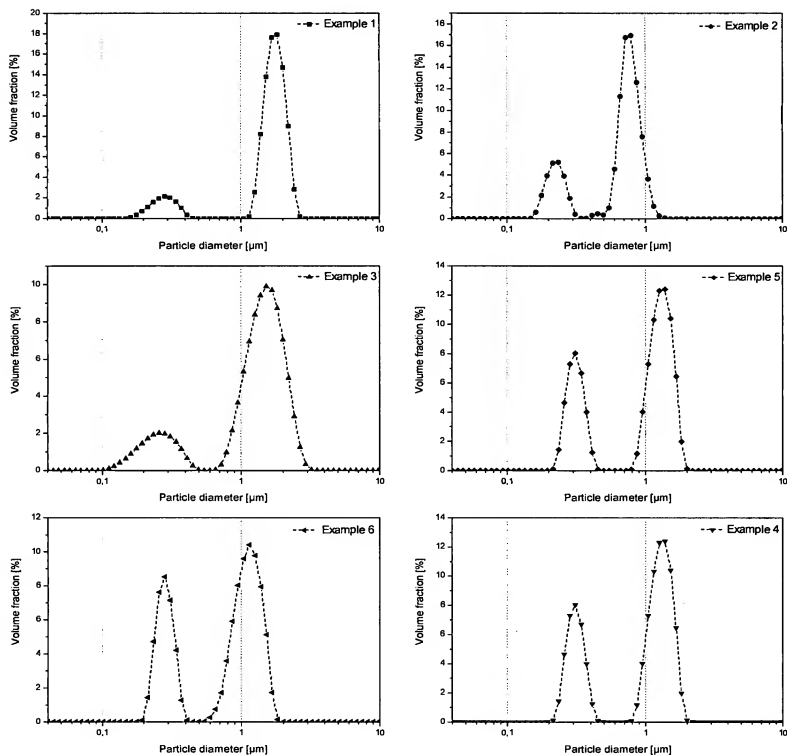


Table 1 (numerical data of examples 1-6):

Particle diameter [μm]	Example 1		Example 2		Example 3		Example 4		Example 5		Example 6	
	Volume fraction [%]	Error	Volume fraction [%]	Error	Volume fraction [%]	Error	Volume fraction [%]	Error	Volume fraction [%]	Error	Volume fraction [%]	Error
0.043	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.044	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.048	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.053	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.059	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.064	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.070	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.077	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.084	0.000 ± 0.179		0.000 ± 0.169		0.000 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.093	0.000 ± 0.179		0.000 ± 0.169		0.004 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.102	0.000 ± 0.179		0.000 ± 0.169		0.027 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.112	0.000 ± 0.179		0.000 ± 0.169		0.098 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.122	0.000 ± 0.179		0.000 ± 0.169		0.0230 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.134	0.001 ± 0.179		0.000 ± 0.169		0.0420 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.146	0.013 ± 0.179		0.037 ± 0.169		0.0040 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.182	0.100 ± 0.179		0.0580 ± 0.169		0.0890 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.178	0.0330 ± 0.179		0.02130 ± 0.169		0.15170 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.195	0.090 ± 0.179		0.03940 ± 0.169		0.0450 ± 0.099		0.061 ± 0.124		0.000 ± 0.124		0.100 ± 0.104	
0.214	0.1430 ± 0.179		0.05320 ± 0.169		0.0470 ± 0.099		0.0340 ± 0.124		0.100 ± 0.124		0.0450 ± 0.104	
0.235	0.1860 ± 0.179		0.05190 ± 0.169		0.010 ± 0.099		0.3380 ± 0.124		0.000 ± 0.124		0.030 ± 0.104	
0.259	0.1950 ± 0.179		0.03310 ± 0.169		0.2000 ± 0.099		0.010 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.284	0.2420 ± 0.179		0.1880 ± 0.169		0.1960 ± 0.099		0.7450 ± 0.124		0.000 ± 0.124		0.8520 ± 0.104	
0.311	0.2020 ± 0.179		0.0420 ± 0.169		0.1610 ± 0.099		0.7330 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.342	0.1930 ± 0.179		0.023 ± 0.169		0.1540 ± 0.099		0.5240 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.375	0.040 ± 0.179		0.029 ± 0.169		0.1350 ± 0.099		0.2750 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.412	0.0360 ± 0.179		0.0280 ± 0.169		0.0860 ± 0.099		0.0760 ± 0.124		0.000 ± 0.124		0.089 ± 0.104	
0.452	0.029 ± 0.179		0.0440 ± 0.169		0.0250 ± 0.099		0.050 ± 0.124		0.089 ± 0.124		0.000 ± 0.104	
0.498	0.000 ± 0.179		0.0320 ± 0.169		0.048 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.545	0.000 ± 0.179		0.0090 ± 0.169		0.004 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.598	0.000 ± 0.179		0.4570 ± 0.169		0.003 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.657	0.000 ± 0.179		0.14300 ± 0.169		0.050 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.721	0.000 ± 0.179		0.0700 ± 0.169		0.0310 ± 0.099		0.000 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
0.791	0.000 ± 0.179		0.0600 ± 0.169		0.0980 ± 0.099		0.100 ± 0.124		0.078 ± 0.124		0.000 ± 0.104	
0.869	0.000 ± 0.179		0.12600 ± 0.128		0.2150 ± 0.099		0.1390 ± 0.124		0.1970 ± 0.124		0.000 ± 0.104	
0.953	0.000 ± 0.179		0.7540 ± 0.075		0.3650 ± 0.099		0.2490 ± 0.124		0.4020 ± 0.124		0.000 ± 0.104	
1.047	0.000 ± 0.179		0.3630 ± 0.036		0.5320 ± 0.099		0.7900 ± 0.124		0.7300 ± 0.124		0.000 ± 0.104	
1.149	0.000 ± 0.179		0.040 ± 0.111		0.650 ± 0.099		0.0700 ± 0.124		0.000 ± 0.124		0.000 ± 0.104	
1.261	0.000 ± 0.179		0.0270 ± 0.023		0.6300 ± 0.099		0.2400 ± 0.124		0.2300 ± 0.124		0.000 ± 0.098	
1.385	0.000 ± 0.179		0.088 ± 0.001		0.6410 ± 0.099		0.12100 ± 0.121		0.2400 ± 0.124		0.000 ± 0.079	
1.520	0.000 ± 0.179		0.009 ± 0.000		0.8900 ± 0.099		0.9000 ± 0.098		0.0400 ± 0.104		0.000 ± 0.051	
1.669	0.000 ± 0.179		0.000 ± 0.000		0.080 ± 0.087		0.970 ± 0.057		0.850 ± 0.065		0.000 ± 0.017	
1.837	0.000 ± 0.179		0.000 ± 0.000		0.080 ± 0.087		0.950 ± 0.016		0.990 ± 0.020		0.000 ± 0.001	
2.010	0.000 ± 0.147		0.000 ± 0.000		0.030 ± 0.070		0.100 ± 0.001		0.000 ± 0.001		0.000 ± 0.000	
2.207	0.000 ± 0.090		0.000 ± 0.000		0.4990 ± 0.050		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
2.423	0.000 ± 0.028		0.000 ± 0.000		0.2900 ± 0.029		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
2.680	0.000 ± 0.002		0.000 ± 0.000		0.1530 ± 0.013		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
2.929	0.000 ± 0.000		0.000 ± 0.000		0.0340 ± 0.003		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
3.206	0.000 ± 0.000		0.000 ± 0.000		0.046 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
3.519	0.000 ± 0.000		0.000 ± 0.000		0.002 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
3.862	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
4.241	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
4.656	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
5.111	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
5.611	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
6.156	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
6.751	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
7.421	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
8.147	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
8.944	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
9.819	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
10.780	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	
11.830	0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000		0.000 ± 0.000	

For better visualization, those datapoints, which contribute to the peaks of the comparative examples are highlighted with grey background. The manufacturer of the instruments states a reproducibility of 1%. Therefore a data point is regarded to contribute to the peak of the particle size distribution only, if its value is larger than the typical relative error (1%) of the peak maximum.

It can be stated, that all comparative examples are in accordance with the claims WO 03/078496. The peaks are well separated and do not overlap.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information or belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed at 67056 Ludwigshafen, Germany, this

day of July 18th 2007

Daniel Frolck

Signature of Declarant